Thermoacoustic Properties of Methyl Esters of *n*-Alkanoic Acids

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ABSTRACT: A number of thermoacoustic parameters, including the Sharma constant S_0, the isochoric temperature coefficient of volume expansivity $(d \ln\alpha/d \ln T)_V$, the isochoric temperature coefficient of internal pressure $(d \ln P_i/d \ln T)_V$, reduced volume V, reduced compressibility β and the Huggins parameter F, are estimated by using only the volume expansivity α from the density temperature data for a number of methyl esters of *n*-alkanoic acids. The methyl esters give excellent separation of saturated fatty acid mixtures. The Sharma constant, which relates the molecular constant *n* and other thermoacoustic parameters, is a constant with a characteristic value of 1.11 + 0.01 for all these esters.

JAOCS 72, 687–691 (1995).

KEY WORDS: Alkanoic acids, density, Huggins parameter, internal pressure, isochoric temperature coefficient, methyl esters, reduced compressibility, reduced volume, Sharma constant, thermoacoustic properties, volume expansivity.

The Sharma constant, S_0, relates the isothermal volume derivative of the intermolecular constant to the thermodynamic Gruneisen parameter and various other physical parameters. As such, it is an important parameter in investigating the physicochemical behavior of any system. As per the investigations of Sharma (1–5) and Sharma and Reddy (6,7), the value of the Sharma constant is constant for any system in either the liquid or the solid state. Venkatacharyulu and coworkers (8–10), Subba Rao (11), and Prasad (12) concluded that even for mesogens in their mesomorphic state in which they simultaneously exhibit the rheological properties of liquids and the anisotropic properties of crystals, S_0 is a constant with the same characteristic value of 1.11 + 0.01 as was reported by Sharma (1–5) and by Sharma and Reddy (6,7) for certain nonmesomorphic systems. Venkatacharyulu and coworkers (8–10) observed from their studies on mesogens that S_0 assumes a lower value than the characteristic value only at and in the immediate vicinity of the phase transition temperature when the system transits from an isotropic liquid state to a mesomorphic state (nematic or smectic), i.e., from a completely low-density isotropic liquid to a more ordered (orientationally or positionally) dense state. Exactly at the transition temperature, S_0 drops to the lowest value. Recently, Murthy (13) carried out similar investigations with a number of molecular liquids and aqueous solutions to determine whether these studies will improve our understanding of the fluid structure. Venkatacharyulu and associates (unpublished data) recently extended the Sharma theory to certain vegetable oils and their constituent fatty acids.

In continuation of the investigation on vegetable oils and fatty acids, the authors estimated a number of thermoacoustic parameters, including the Sharma constant, for methyl esters of some *n*-alkanoic acids by using only the coefficient of volume expansion α. Such studies have not been carried out on methyl esters, which are known to give excellent separation of fatty acids. These parameters may be useful in the fatty acid industry to characterize fatty acids and to help understand the liquid structure of these esters. The coefficients of volume expansion are estimated from Liew *et al.*'s (14) density reports.

RESULTS AND DISCUSSION

All acoustic parameters under consideration are estimated from the volume expansivity α, which can be written as:

$$\alpha = \frac{1}{V_n}\frac{\Delta V}{\Delta T} \qquad [1]$$

where $V_n = (V_1 + V_2)/2$, $\Delta V = (V^2 - V_1)$, $\Delta T = (T_2 - T_1)$. V_2 and V_1 are the molar volumes at temperatures T_2 and T_1, respectively. The values of this parameter for all methyl esters are given in Tables 1–5.

Isochoric temperature coefficient of internal pressure. Sharma obtained an expression for the isochoric temperature coefficient of internal pressure by using the coefficient of thermal expansion α as:

$$\left[\frac{d \ln P_i}{d \ln T}\right]_V = \frac{2}{\beta}\left[\frac{d \ln \alpha}{d \ln T}\right]_V = \frac{-2(1+2\alpha T)}{\left(\tilde{V}\right)^{C_i}} \qquad [2]$$

where P_i is the internal pressure, and β, $\tilde{V}$, and C_1 are as defined later. Tables 1–5 readily show that the coefficient of in-

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TABLE 1
Thermoacoustic Parameters of Methyl Ester of C_6 *n*-Alkanoic Acid

T (K)	$\alpha \times 10^2$	$\left[\frac{d \ln \alpha}{d \ln T}\right]_V$	$\left[\frac{d \ln P_i}{d \ln T}\right]_V$	β	*V*	S_0	*S**	S_0^*	*F*
283	0.1170	1.6621	0.5161	6.4407	1.2699	1.1159	1.4414	1.1531	1.0747
288	0.0950	1.5467	0.5469	5.6555	1.2304	1.1195	1.3645	1.1335	1.1824
293	0.1205	1.7061	0.5049	6.7577	1.2842	1.1139	1.4707	1.1600	1.0341
298	0.1189	1.7089	0.5042	6.7783	1.2851	1.1138	1.4726	1.1604	1.0316
303	0.1080	1.6544	0.5180	6.3868	1.2673	1.1162	1.4363	1.1518	1.0817
308	0.1063	1.6548	0.5180	6.3891	1.2674	1.1162	1.4365	1.1519	1.0814
313	0.1163	1.7278	0.4995	6.9177	1.2912	1.1129	1.4852	1.1633	1.0143
318	0.2081	2.3235	0.3756	12.3705	1.4534	1.0606	1.8823	1.2343	0.4933
323	0.1152	1.7445	0.4954	7.0426	1.2965	1.1120	1.4963	1.1658	0.9990
328	0.0945	1.6199	0.5271	6.1460	1.2557	1.1174	1.4132	1.1462	1.1138
333	0.1236	1.8235	0.4766	7.6523	1.3209	1.1074	1.5490	1.1772	0.9275
338	0.1172	1.7923	0.4839	7.4073	1.3114	1.1093	1.5282	1.1728	0.9557
343	0.1106	1.7589	0.4919	7.1513	1.3010	1.1112	1.5059	1.1679	0.9859
349	0.1332	1.9268	0.4533	8.5010	1.3512	1.1001	1.6179	1.1909	0.8354

ternal pressure $(d \ln P_i/d \ln T)_V$ is a negative quantity with a value around 0.5 at all temperatures for all methyl esters under consideration, which is in accordance with the observations made by earlier investigators (4,7–10) for systems other than methyl esters. This is true for all methyl esters under consideration except when the number of carbon atoms is C = 7 at 318 K, where $(d \ln P_i/d \ln T)_V$ exhibits a huge negative value. This large change in the isochoric temperature coefficient of internal pressure is due to the high value in the volume expansivity at this particular temperature when compared to any other temperature for all homologous methyl esters under investigation. At 318 K for the methyl ester with C = 7, α exhibits anomalous behavior. Hence, it can be concluded that $(d \ln P_i/d \ln T)_V$ is a constant for a particular system, independent of temperature and depending on α alone. Thus, the sudden change in $(d \ln P_i/d \ln T)_V$ in the methyl ester with C = 7 is due to the sudden change in α, as $(d \ln P_i/d \ln T)_V$ is estimated only from α. Similar conclusions were drawn by Murthy (13) for a number of molecular liquids.

Isochoric temperature coefficient of volume expansion. This parameter for all members of the homologous series of methyl esters of *n*-alkanoic acids under investigation (C = 6, 7, 8, 10, and 12) are presented in Tables 1–5. The isochoric temperature coefficient of volume expansivity can be obtained from:

$$\left[\frac{d \ln \alpha}{d \ln T}\right]_V = -S^* S_0^* = -(1 + 2\alpha T) \qquad [3]$$

where S^* and S_0^* are the Sharma parameters defined below.

C_1, the pressure coefficient of bulk modules, introduced by Moelwyn-Hughes (16), can be written as:

$$C_1 = \left[\frac{d}{dp}\left(\frac{1}{\beta}\right)\right] = \left[\frac{d \ln \beta}{d \ln V}\right]_T \qquad [4]$$

Sharma (16) obtained the compressibility expression from the reduced equation of state to estimate C_1 and calculated

TABLE 2
Thermoacoustic Parameters of Methyl Ester of C_7 *n*-Alkanoic Acid

T (K)	$\alpha \times 10^2$	$\left[\frac{d \ln \alpha}{d \ln T}\right]_V$	$\left[\frac{d \ln P_i}{d \ln T}\right]_V$	β	*V*	S_0	*S**	S_0^*	*F*
283	0.1017	1.5759	0.5396	5.8478	1.2406	1.1188	1.3839	1.1387	1.1550
288	0.1023	1.5890	0.5353	5.9360	1.2452	1.1184	1.3927	1.1409	1.1426
293	0.1051	1.6158	0.5282	6.1179	1.2543	1.1176	1.4105	1.1455	1.1176
298	0.1056	1.6296	0.5245	6.2130	1.2590	1.1171	1.4197	1.1478	1.1048
303	0.1103	1.6717	0.5136	6.5093	1.2730	1.1155	1.4478	1.1546	1.0658
308	0.1044	1.6431	0.5210	6.3069	1.2635	1.1166	1.4287	1.1500	1.0923
313	0.1097	1.6867	0.5098	6.6172	1.2780	1.1148	1.4578	1.1570	1.0519
318	0.9852	7.2660	0.0372	390.2190	1.9657	0.2889	5.1773	1.4034	3.1401
323	0.1061	1.6854	0.5101	6.6077	1.2775	1.1149	1.4569	1.5680	1.0531
328	0.1091	1.7154	0.5026	6.8259	1.2872	1.1135	1.4769	1.1614	1.0256
333	0.1265	1.8422	0.4722	7.8011	1.3265	1.1062	1.5614	1.1797	0.9108
338	0.1038	1.6970	0.5072	6.6912	1.2813	1.1143	1.4647	1.1586	1.0426
343	0.1134	1.7776	0.4874	7.2934	1.3069	1.1102	1.5184	1.1707	0.9690
348	0.1164	1.8104	0.4796	7.5484	1.3169	1.1082	1.5402	1.1753	0.9393

TABLE 3
Thermoacoustic Parameters of Methyl Ester of C_8 *n*-Alkanoic Acid

T (K)	$\alpha \times 10^2$	$\left[\frac{d \ln \alpha}{d \ln T}\right]_V$	$\left[\frac{d \ln P_i}{d \ln T}\right]_V$	β	*V*	S_0	*S**	S_0^*	*F*
283	0.0975	1.5521	0.5454	5.6908	1.2323	1.1194	1.3681	1.1345	1.1773
288	0.1003	1.5779	0.5384	5.6110	1.2413	1.1188	1.3852	1.1390	1.1531
293	0.1054	1.6178	0.5276	6.1315	1.2550	1.1175	1.4119	1.1458	1.1157
298	0.1037	1.6179	0.5276	6.1323	1.2550	1.1175	1.4119	1.1458	1.1157
303	0.0972	1.5892	0.5353	5.9369	1.2452	1.1184	1.3928	1.1410	1.1426
308	0.1024	1.6307	0.5242	6.2207	1.2594	1.1171	1.4204	1.1480	1.1038
313	0.1029	1.6442	0.5207	6.3151	1.2639	1.1166	1.4295	1.1502	1.0912
318	0.1034	1.6579	0.5171	6.4112	1.2685	1.1160	1.4386	1.1524	1.0785
323	0.1064	1.6871	0.5097	6.6198	1.2781	1.1148	1.4581	1.1570	1.0516
328	0.1045	1.6857	0.5100	6.6094	1.2776	1.1149	1.4571	1.1568	1.0530
333	0.1027	1.6839	0.5105	6.5965	1.2770	1.1149	1.4559	1.1565	1.0546
338	0.1105	1.7457	0.4948	7.0586	1.2972	1.1119	1.4978	1.1661	0.9970
343	0.1111	1.7620	0.4911	7.1743	1.3020	1.1110	1.5080	1.1684	0.9831
348	0.1111	1.7944	0.4834	7.4233	1.3120	1.1092	1.5296	1.1731	0.9538

the values of β and α according to the expressions.

$$\beta = \alpha T \tilde{V}^2 / P^* \qquad [5]$$

$$\alpha = 3\left(\tilde{V}^{1/3} - 1\right) / \left(4 - 3\tilde{V}^{1/3}\right) \qquad [6]$$

where $\tilde{V} = V/V^*$ is the reduced volume, V is the molar volume, V^* is the characteristic volume, and P^* is the characteristic pressure. However, for the present work, α was estimated from Equation 1.

Tables 1–5 show that this parameter is independent of temperature and is dependent on the volume expansivity α only. It decreases with the increase of α and is almost a constant with a value of around −1.6 for this series, except for the ester with C = 7 at 318 K where α exhibits anomalous behavior. These results are in accordance with the results in different systems investigated earlier (4,7–10).

Reduced volume V. The reduced volume $\tilde{V}$, a function of the volume expansivity and temperature in Equation 6, is:

$$\tilde{V} = \left[\frac{1 + \alpha T}{3(1 + \alpha T)}\right] \qquad [7]$$

Tables 1–5 show that the values of V for the five methyl esters under investigation are almost constant with values of 1.3 + 0.01, except for C = 7 at 318 K, where this parameter has a large value, 1.97. The value of V for all other systems investigated by Sharma (4), Sharma and Reddy (7), and Venkatacharyulu and colleagues (8–10) was in the range of 1.0 to 1.412, and thus, the values for the methyl esters are comparable to other nonmesomorphic and mesomorphic systems, including certain vegetable oils recently investigated by Subramanyam, M.S.R., H. Sumathi Vedanayagam, and P. Venkatacharyalu (unpublished data).

Sharma used Equations 4, 5, and 6 to estimated C_1 in terms of only α and T.

TABLE 4
Thermoacoustic Parameters of Methyl Ester of C_{10} *n*-Alkanoic Acid

T (K)	$\alpha \times 10^2$	$\left[\frac{d \ln \alpha}{d \ln T}\right]_V$	$\left[\frac{d \ln P_i}{d \ln T}\right]_V$	β	*V*	S_0	*S**	S_0^*	*F*
283	0.0981	1.5554	0.5445	5.7124	1.2334	1.1193	1.3703	1.1351	1.2092
288	0.0875	1.5154	0.5558	5.4527	1.2192	1.1202	1.3436	1.1278	1.1856
293	0.0967	1.5669	0.5414	5.7879	1.2375	1.1191	1.3779	1.1371	1.1614
298	0.0949	1.5655	0.5418	5.7786	1.2370	1.1191	1.3770	1.1369	1.2021
303	0.0977	1.5919	0.5346	5.9551	1.2462	4.4483	1.3946	1.1414	1.1513
308	0.0958	1.5901	0.5350	5.9430	1.2456	1.1184	1.3934	1.1411	1.1943
313	0.0963	1.6026	0.5317	6.0279	1.2498	1.1180	1.4017	1.1433	1.1560
318	0.0991	1.6303	0.5243	6.2182	1.2593	1.1171	1.4202	1.1479	1.1025
323	0.0924	1.5972	0.5331	5.9915	1.2480	1.1182	1.3981	1.1423	1.1334
328	0.1048	1.6878	0.5095	6.6245	1.2783	1.1148	1.4585	1.1572	1.1368
333	0.0958	1.6378	0.5224	6.2699	1.2618	1.1168	1.4252	1.1491	1.0959
338	0.1035	1.6996	05065	6.7101	1.2821	1.1142	1.4664	1.1590	1.0841
343	0.1016	1.6969	0.5072	6.6905	1.2813	1.1143	1.4646	1.1586	1.0568
348	0.1095	1.7618	0.4912	7.1727	1.3019	1.1111	1.5078	1.1684	1.0288

TABLE 5
Thermoacoustic Parameters of Methyl Ester of C_{12} *n*-Alkanoic Acid

T (K)	$\alpha \times 10^2$	$\left[\frac{d\ln\alpha}{d\ln T}\right]_V$	$\left[\frac{d\ln P_i}{d\ln T}\right]_V$	β	*V*	S_0	*S**	S_0^*	*F*
282	0.0916	1.5185	0.5549	5.4728	1.2203	1.1201	1.3457	1.1284	1.2092
288	0.0943	1.5434	0.5479	5.6334	1.2292	1.1196	1.3623	1.1329	1.1856
293	0.0971	1.5691	0.5408	5.8026	1.2382	1.1190	1.3794	1.1375	1.1614
298	0.0883	1.5260	0.5528	5.5208	1.2230	1.1200	1.3506	1.1298	1.2021
303	0.0957	1.5798	0.5379	5.8738	1.2420	1.1187	1.3865	1.1393	1.1513
308	0.0867	1.5342	0.5505	5.5736	1.2259	1.1198	1.3561	1.1313	1.1943
313	0.0918	1.5748	0.5392	5.8407	1.2402	1.1188	1.3832	1.1385	1.1560
318	0.0994	1.6320	0.5239	6.2301	1.2598	1.1170	1.4213	1.1482	1.1025
323	0.0927	1.5989	0.5327	6.0026	1.2486	1.1181	1.3992	1.1426	1.1334
328	0.0907	1.5953	0.5337	5.9780	1.2473	1.1182	1.3968	1.1420	1.1368
333	0.0960	1.6392	0.5220	6.2798	1.2622	1.1168	1.2461	1.1494	1.0959
338	0.0964	1.6519	0.5187	6.3687	1.2665	1.1163	1.4346	1.1514	1.0841
343	0.0993	1.6815	0.5111	6.5792	1.2762	1.1150	1.4543	1.1562	1.0568
348	0.1038	1.7119	0.5034	6.8001	1.2861	1.1136	1.4746	1.1609	1.0288

$$C_1 = \left[\frac{13}{3} + (\alpha T)^{-1} + \frac{4}{3}\alpha T\right] \qquad [8]$$

It can be concluded that α is the controlling factor of C_1 and an important parameter to estimate the temperature dependence of C_1.

Reduced compressibility β. The isochoric temperature coefficient of compressibility β can be obtained from Equations 2 and 3 as:

$$\left[\frac{d\ln\beta}{d\ln T}\right] = \left[1 - 2\alpha T\left(\tilde{V}^{C_1} - 1\right)\right] \Big/ \left(\tilde{V}\right)^{C_1} + 2\alpha T \qquad [9]$$

and the reduced compressibility (7) is:

$$\beta = \tilde{V}^{C_1} \qquad [10]$$

As it can be observed from Tables 1–5, β is estimated as a function of temperature for all five methyl esters at normal atmospheric pressure, and all values lie mostly between 5 and 12, except for the methyl ester with C = 7 at 318 K, where the value of this parameter is as high as 390.62. Unlike other parameters discussed earlier, this parameter is more sensitive to the volume expansivity, i.e., the higher the α the higher the β.

The Sharma constant S_0. Sharma expressed S_0 as:

$$S_0 = -\frac{1}{2}\left(\frac{d\ln P_i}{d\ln T}\right)_V \left(\frac{\alpha}{V}\right) = \frac{S^* S_0^* \alpha}{\beta V} = \frac{(1+2\alpha T)}{\tilde{V}^{C_1}}(3+4\alpha T) \qquad [11]$$

If the parameters α, S^*, and S_0^* are expressed in terms of α only, Sharma obtained:

$$S^* = \frac{\tilde{\alpha}}{3V} = 1 + \frac{4}{3}\alpha T = 1 + \frac{4}{3}\alpha T \qquad [12]$$

$$S_0^* = -\left(\frac{d\ln\alpha}{d\ln T}\right)_V \Big/ S^* = \frac{1+2\alpha}{1+\frac{4}{3}\alpha T} \qquad [13]$$

$$\bar{\alpha} = \left(\frac{d\ln\tilde{V}}{dT}\right)_P = \alpha T^* \qquad [14]$$

$$\tilde{T} = T \Big/ T^*\left(V^{\frac{1}{3}} - 1\right) \Big/ \tilde{V}^{\frac{4}{3}} \qquad [15]$$

$$\frac{\tilde{V}^{\frac{1}{3}}}{\tilde{V}^{\frac{4}{3}}} = \left(\tilde{V}\tilde{T}\right)^{-1} = \frac{(3+4\alpha T)}{\alpha T} = \frac{3S^*}{\tilde{\alpha}\tilde{T}} = \frac{3S^*}{\alpha T} \qquad [16]$$

where $\tilde{\alpha}$ represents the reduced volume expansivity, $\tilde{T}$ is the reduced temperature, and T^* is the characteristic temperature of the substance.

Parameter S.* The estimated values of S^* for the five methyl esters under investigation are presented in Tables 1 to 5, and they are constant with a value of 1.5 ± 0.02, except for the methyl ester with C = 7 at 318 K, where α exhibits anomalous behavior. Again, the value of S^* for polymers reported by Sharma and Reddy (7) are in the range of 1.06 to 1.62, for alkali halides they are around 1.5, for polycrystalline solids it is 1.2, for ionic liquids they range from 1.179 to 1.644, and for liquid crystals Venkatacharyulu *et al.* (8–10) reported a value of 1.44.

Huggins parameter F. Huggins (17,18) developed a parameter that may be expressed in terms of temperature and pressure derivatives of compressibility as:

$$F = 1 + \frac{2}{3}\alpha T + T\left(\frac{d\ln\beta}{dT}\right)_P + \left(\frac{\alpha T}{\beta}\right)\left(\frac{d\ln\beta}{dP}\right)_T \qquad [17]$$

If the compressibility is assumed to be a function of only volume and temperature, according to Sharma thermodynamic considerations show that:

$$\frac{\alpha}{\beta}\left(\frac{d\ln\beta}{dP}\right)_T + \left(\frac{d\ln\beta}{dT}\right)_P = \left(\frac{d\ln\beta}{d\ln T}\right)_V \qquad [18]$$

From Equations 9, 10, 17, and 18, one can easily obtain:

$$F = 1 + \frac{2}{3}\alpha T + T\left(\frac{d\ln\beta}{dT}\right)_V = 1 + \frac{2}{\tilde{V}^{C_1}} + (4\alpha T)\left[\frac{2}{3} - \frac{\left(\tilde{V}^{C_1} - 1\right)}{\tilde{V}^{C_1}}\right] \quad [19]$$

From Equations 11–13 and 19, the Huggins parameter, F, can also be expressed in terms of the Sharma constant S_0 and S_0^*. All parameters can again be expressed in terms of one single measurable parameter, α:

$$F = 2\left[1 + \frac{S_0}{3S^*}\right] - S^* = 2\left(1 + \frac{\alpha T}{3}\right) - S_0^* + \frac{2S_0}{3S^*}$$

$$= 1 - \frac{2\alpha T}{3} - S^*(S_0^* - 1) + \frac{2S_0}{3S^*} \quad [20]$$

By using Equation 13, S_0 can also be expressed as:

$$S_0 = \left(\frac{d\ln\alpha}{d\ln T}\right)_V \frac{\tilde{V}_0}{V\tilde{V}^2} = \frac{S^* S_0^* \tilde{V}_0}{\tilde{V}^2} \quad [21]$$

where $\tilde{V}_0 = \tilde{\alpha}\tilde{V}/\beta$.

It is evident from these equations that the coefficient of volume expansivity α is an important parameter, which can be used to estimate various physical parameters S_0, S^*, S_0^*, and F. The authors have successfully applied Sharma's theory and obtained the various thermoacoustic parameters for the five methyl esters of *n*-alkanoic acids reported in the present studies.

S_0, which is estimated as a function of temperature for all five methyl esters studied, is independent of temperature and dependent only on α. As the volume expansivity α increases, S_0F decreases. For all five methyl esters investigated, the volume expansivity is almost of the same order, but α decreases with the increasing number of carbon atoms. The Sharma constant, which relates the isothermal volume derivative of intermolecular constant to the thermodynamic Gruneisen parameter and other various physical parameters, should be constant and independent of the number of carbon atoms in the homologous series. Even though the intermolecular constant and the thermodynamic Gruneisen parameters are specific to that liquid and vary from liquid to liquid, the Sharma constant is the same for all members of the series.

In only one case, namely, the methyl ester of heptanoic acid at 318 K, does the Sharma constant assume a low value (as low as 0.289) due to the high value of the volume expansivity at this temperature. Such instances were reported by Venkatacharyulu and co-workers (8–10) in their application of Sharma's theory to liquid crystals. They observed that the Sharma constant falls to a low value at the transition temperature when the mesogen transits from one mesophase to the other or from a highly disordered low density state to a more ordered (positional order or orientation order) high density state. Thus, it can be concluded that the molecules in the liquid may themselves become arranged in an orderly way. The variation in density at this particular temperature is slightly higher when compared to the other temperatures in this ester. The reason for this can be understood more fully by further studies that include differential scanning calorimetry.

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[Received June 7, 1994, accepted December 19, 1994]